# New Mechanisms and Materials for Odd-Frequency Superconductivity

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Hybrid Correlated States and Dynamics in Quantum Materials
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# Outline

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- Introduction to odd-frequency ($\omega$) superconductivity
  - What and where?
- Weyl nodal loop semimetals
  - "Optimal" odd-$\omega$ system
- Odd-$\omega$ pairing in multiband systems
  - Odd-$\omega$ pairing is ubiquitous in SCs
- QPI as direct probe of odd-$\omega$ pairing

# Acknowledgements


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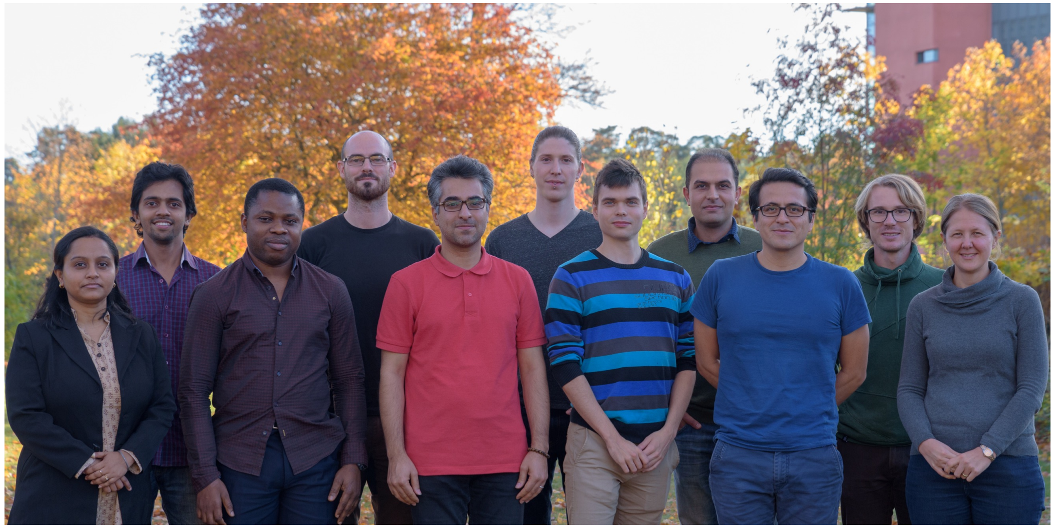


Jorge Cayao, Debmalya Chakraborty, Paramita Dutta, Lucia Komendova, Dushko Kuzmanovski, Tomas Löthman, Fariborz Parhizgar, Johann Schmidt, Andreas Theiler, Christopher Triola

Alexander Balatsky (Nordita), Yukio Tanaka (Nagoya), Jacob Linder (NTNU), Jonas Fransson (Uppsala)

# Odd-Frequency Superconductivity

What and where?

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# Superconductivity

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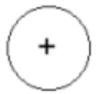
Ordered states → symmetry of order parameter, $\Delta$

Superconductivity: $\Delta \sim F = \langle \psi_\alpha \psi_\beta \rangle$ (Expectation value of forming a Cooper pair)

Fermi-Dirac statistics:

- spin-singlet, *s*-wave
  $\uparrow\downarrow - \downarrow\uparrow$




- spin-triplet, *p*-wave
  $\uparrow\downarrow + \downarrow\uparrow$
  $\uparrow\uparrow, \downarrow\downarrow$

# Superconductivity

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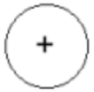
Ordered states → symmetry of order parameter, $\Delta$

Superconductivity: $\Delta \sim F = \langle \psi_\alpha(t)\psi_\beta(0) \rangle$ ($t \leftrightarrow \omega$, frequency)

Cooper pair symmetries

- even-ω, spin-singlet, *s*-wave

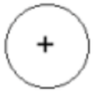
  $\uparrow\downarrow - \downarrow\uparrow$




- **odd-ω**, spin-triplet, *s*-wave

  $\uparrow\downarrow + \downarrow\uparrow$

  $\uparrow\uparrow, \downarrow\downarrow$



# Brief History

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## Bulk, intrinsic phase:

- 1974, Berezinskii: spin-triplet *s*-wave phase in $^3$He
- 1991, Kirkpatrick,Belitz: Disorder-induced spin-triplet *s*-wave SC
- 1992, Balatsky,Abrahmas: spin-singlet *p*-wave SC
- 1994-5, Coleman,Miranda,Tsvelik: Heavy fermions compounds, Majorana fermions
- Complicated models (e.g. composite condensates) and questions about stability (e.g. paramagnetic Meissner effect)

## In this talk:

- Conventional order parameter and induced odd-$\omega$ pairs (correlations)
  E.g. heterostructures or multiband systems

Two reviews: Bergeret, Volkov, Efetov, RMP 77, 1321 (2005), Balatsky and Linder, RMP 91, 045005 (2019)

# SF Interface

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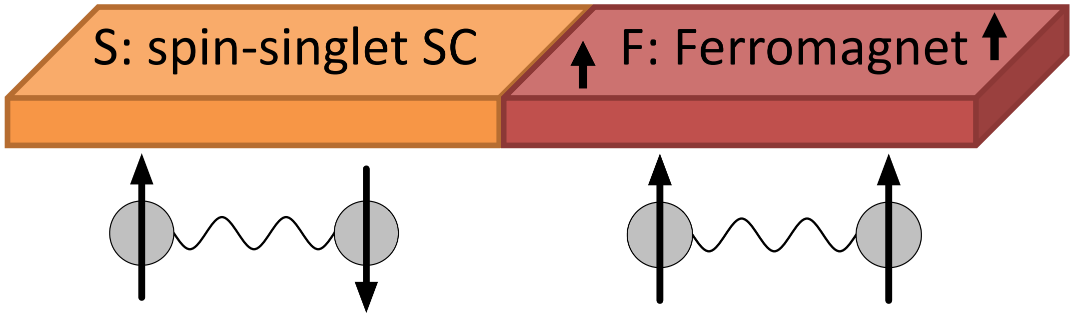


Spin-singlet *s*-wave SC → **odd-ω** spin-triplet *s*-wave pairing

- Long-range SC proximity effect into F → spin-triplet
- Diffusive F → *s*-wave

Bergeret et al, PRL 86, 3140 (2001), Eschrig and Löfwander, Nat. Phys. 4, 138 (2007)

# Physics at the SF Interface

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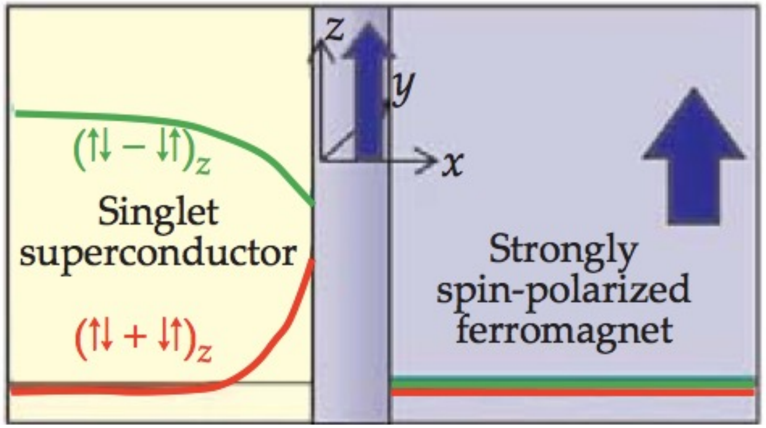


Spin-singlet → Mixed-triplet
~ FFLO physics

Spin-singlet → Mixed-triplet → Equal-triplet
Non-collinear magnetization:

- Two different magnetic layers
- Spin-orbit coupled interface
- Helical ferromagnet

**Diffusive system: spin-triplet & *s*-wave → odd-ω**

Eschrig, Phys. Today 64, 43 (2011)

# Weyl Nodal Loop Semimetals

"Optimal" odd-$\omega$ Josephson junction

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# Weyl/Dirac semimetals

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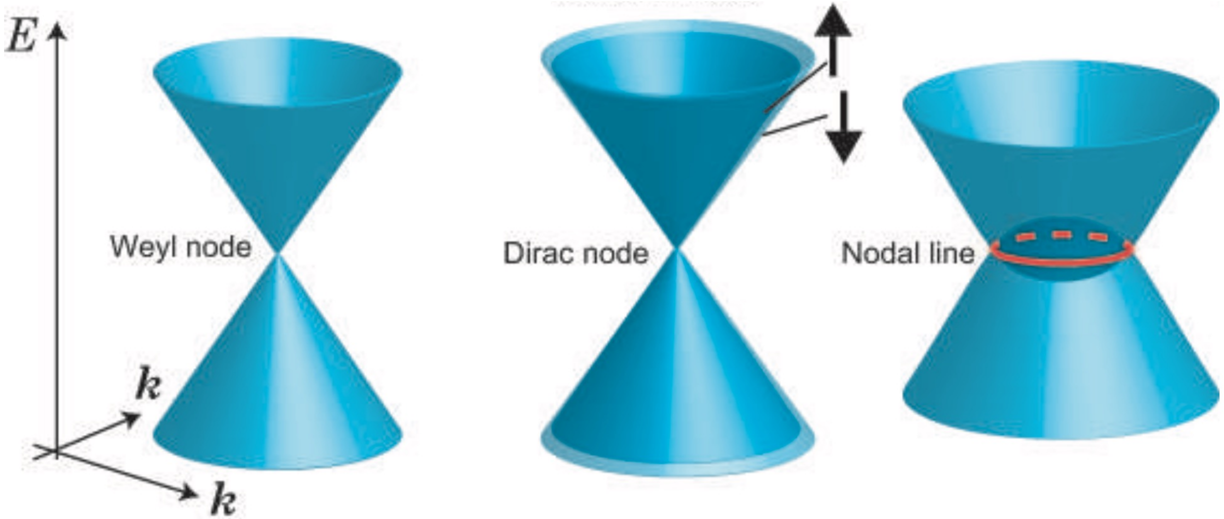
3D bulk materials

Weyl semimetal | Dirac semimetal | **Nodal loop semimetal**

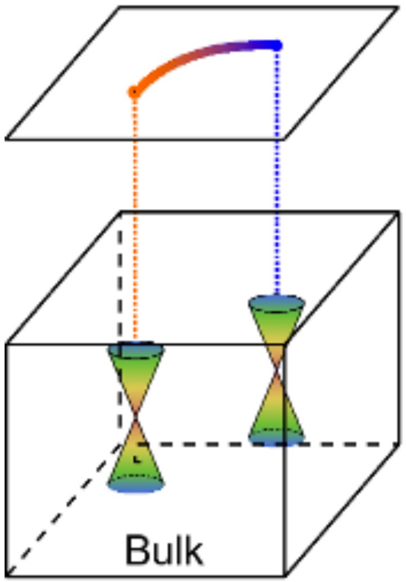



Wehling, ABS, Balatsky, Adv. Phys. 63, 1 (2014), Hirayama et al, JPSP 87, 041002 (2018)

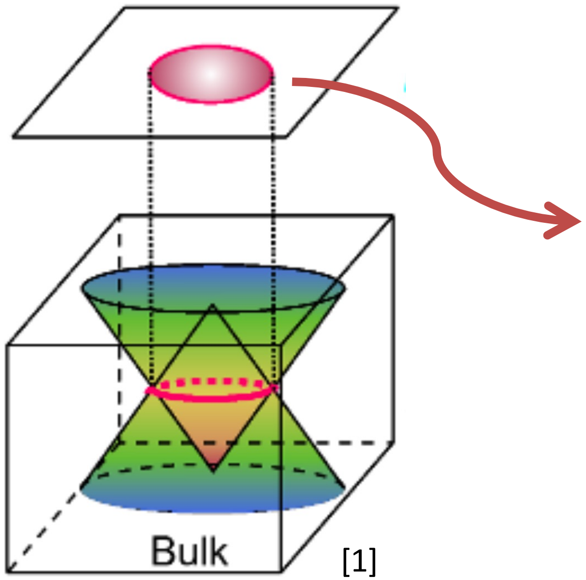
# Surface States

Fermi arc

**Flat drumhead surface state (DSS)**



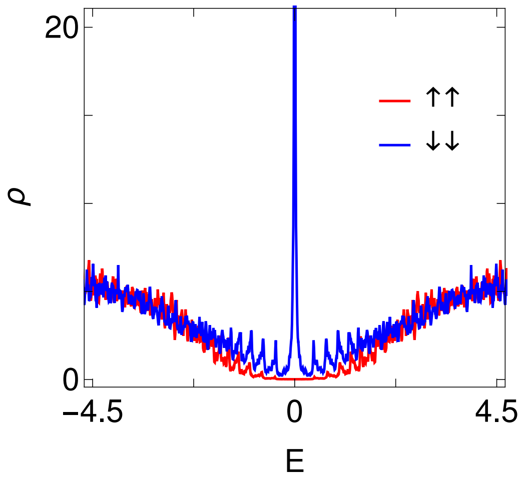

Weyl semimetal




[1]

Weyl nodal loop semimetal (WNL)

DOS on top surface




Proposed materials:
$HgCr_2Se_4$, $PbTaSe_2$,…[2]

[1]: Li et al, Sci. China Matter 62, 23 (2018), [2]: Chen et al, PRL 121, 166802 (2018), Bian et al, Nat. Commun. 7, 10556 (2016)

# Weyl Nodal Loop Semimetal

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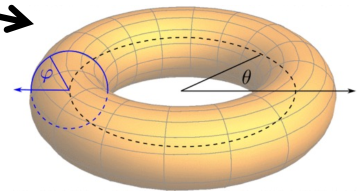
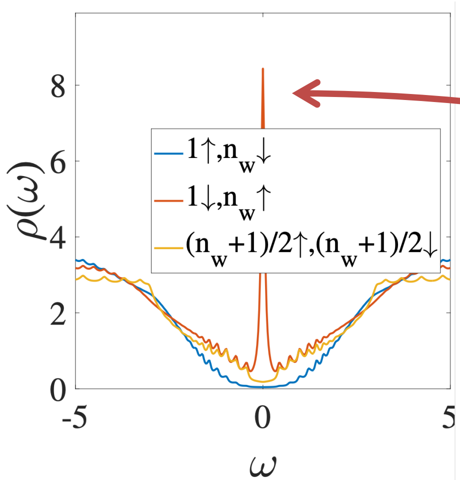
$$H = t_w \left( (\alpha_1 - k^2)\sigma_x + 2\alpha_2 k_z \sigma_y \right) - \mu_w$$

σ in spin-space → **SOC in bulk**




Line or donut
Fermi surface,
α sets shape

DOS per layer in slab




**Spin-polarized DSS**

# Proximity-Induced Superconductivity

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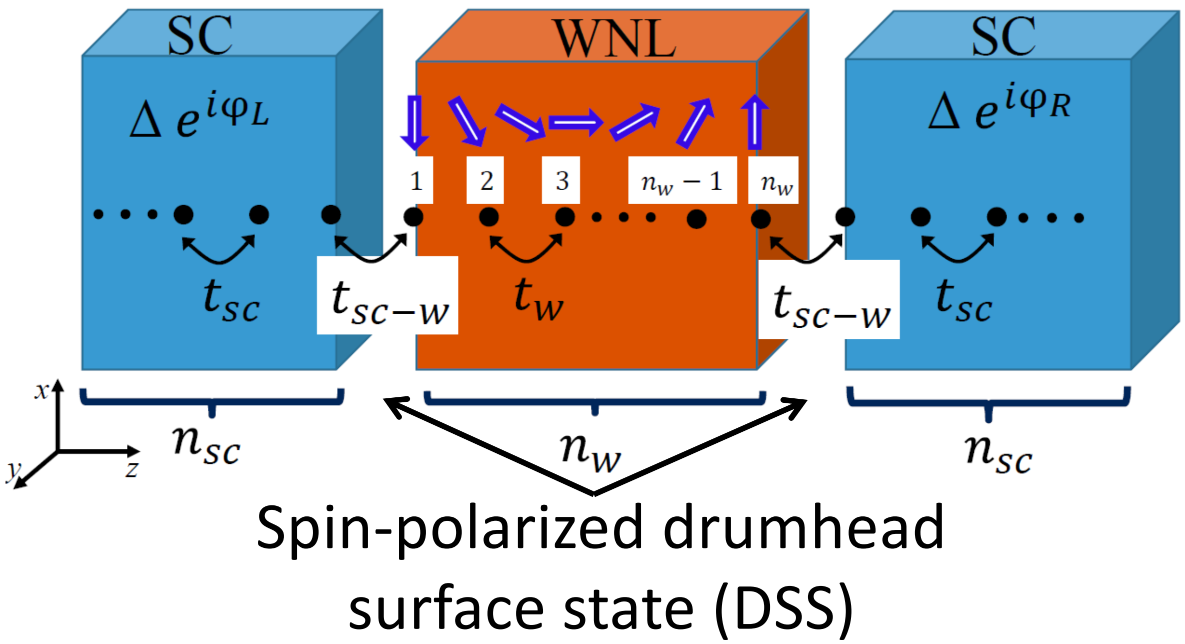
Fully spin-polarized surface → No spin-singlet pairs

**→ No Josephson effect using conventional SCs ?**




Conventional SCs
(spin-singlet *s*-wave)

$n_w = 21$
$n_{sc} = 20$
$t_w = t_{sc} = 1$
$t_{sc\text{-}w} = 0 - 1$
$\Delta = 0.01 - 0.05$

# Superconducting Pairing

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Green's functions

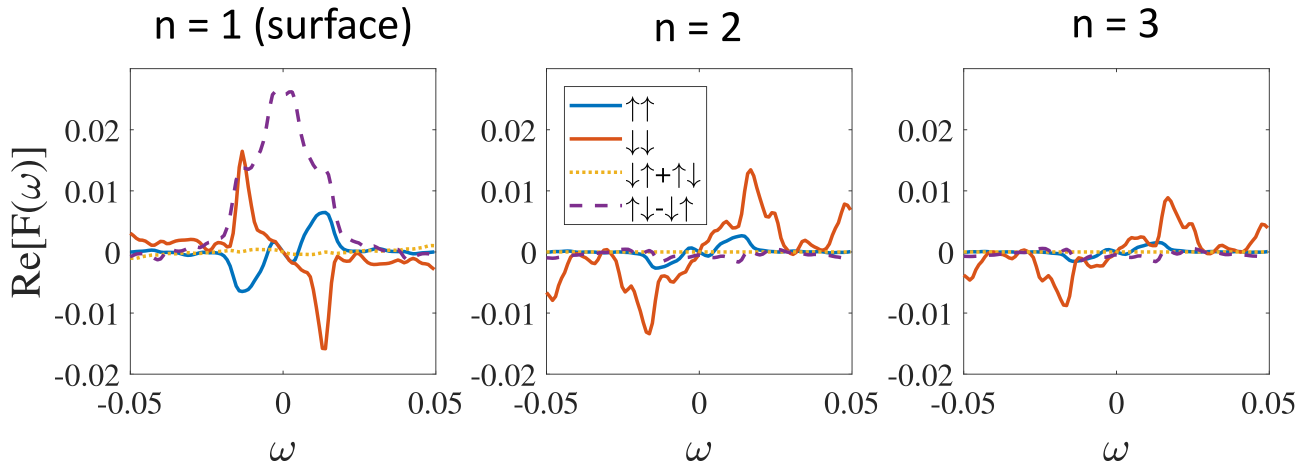
$$G = \left(\omega + i0^{+} - H\right)^{-1}$$

$$G = \begin{pmatrix} g & F \\ \bar{F} & \bar{g} \end{pmatrix}$$

Pair amplitudes:

- Singlet *s*-wave
- Odd-ω triplet *s*-wave
- Negligible *p*-waves

## *s*-wave pair amplitudes




**Only odd-ω triplet *s*-wave pairing survives**

Parhizgar and ABS, npj Quantum Mater. 5, 42 (2020)

# Josephson Current

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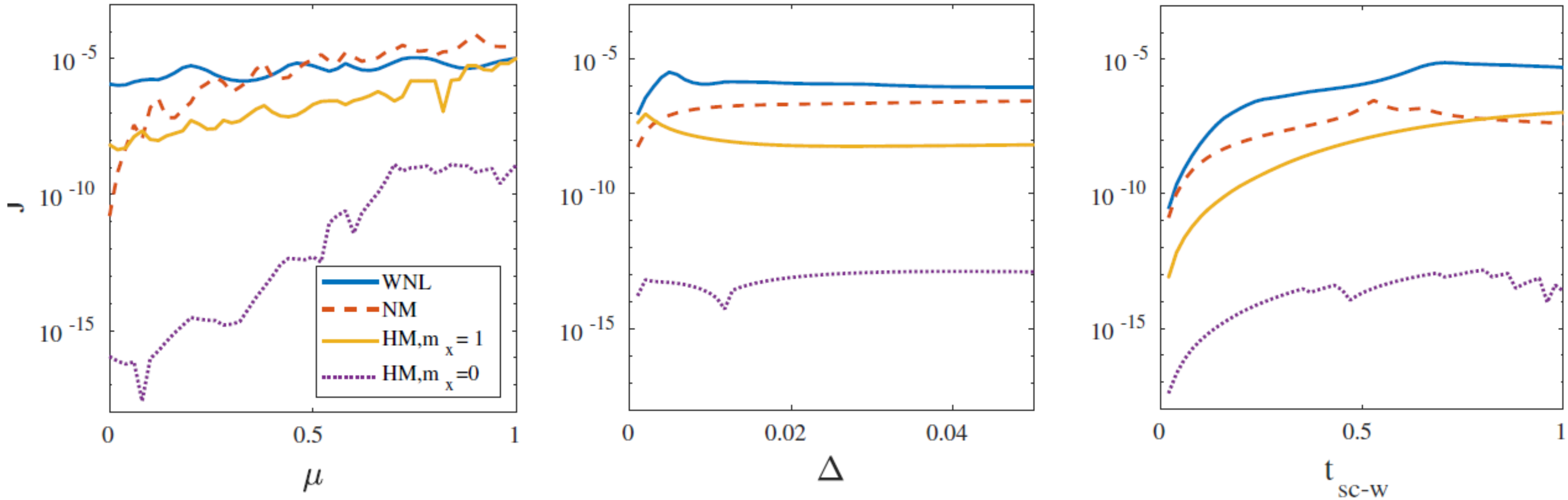


Normal metal (NM); Half-metal (HM): No proximity effect; Non-collinear HM: Odd-ω triplet

WNL: Huge current (>> NM, HM junctions)
due to **flat drumhead surface states**

Parhizgar and ABS, npj Quantum Mater. 5, 42 (2020)

# Odd-ω Pairing in WNLs Junctions

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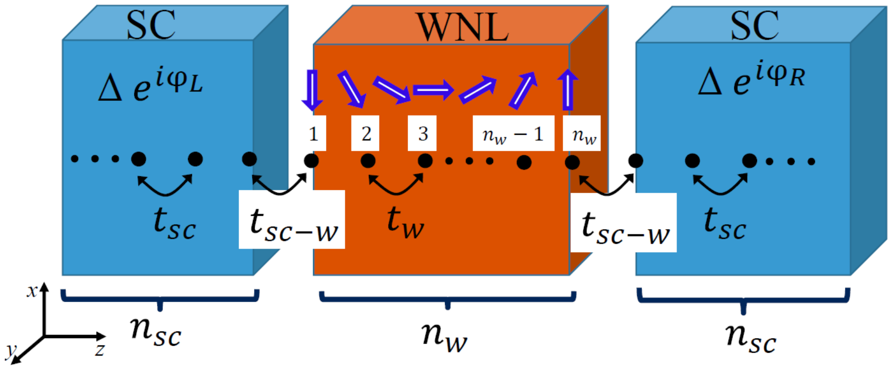


- Spin-polarized flat drumhead surface states & bulk SOC
  - → Only odd-ω spin-triplet *s*-wave pairs
  - → Huge Josephson current

**"Optimal" odd-ω Josephson junction**

Parhizgar and ABS, npj Quantum Mater. 5, 42 (2020)

# Odd-ω Bulk Multiband Superconductivity

Simple two-band SC

Extension to other systems

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# Multiband Superconductors

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Multiple bands (orbitals) → add band index

$$\Delta \sim F = \langle \psi_\alpha(t)\psi_\beta(0)\rangle$$

Cooper pair symmetries

- odd-ω, odd-band, spin-singlet, *s*-wave
- ω ⟷ band $\uparrow\downarrow + \downarrow\uparrow$ (+)

ABS and Balatsky, PRB 88, 104514 (2013)

# Multiband Superconductors

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- S: Spin (even: spin-triplet; odd: spin-singlet)
- P: Spatial parity (even: *s*,*d*-wave; odd: *p*,*f*-wave)
- O: Orbital or band parity (even; odd orbital)
- T: Time (even; odd-frequency)

SPOT = -1

Conventional superconductivity

Odd-ω equivalent

# Simple Two-Band Superconductor

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$$H = \sum_{k\sigma} \varepsilon_1(k) a^\dagger_{k\sigma} a_{k\sigma} + \varepsilon_2(k) b^\dagger_{k\sigma} b_{k\sigma}$$
$$+ \sum_{k} \Delta_1(k) a^\dagger_{k\uparrow} a^\dagger_{-k\downarrow} + \Delta_2(k) b^\dagger_{k\uparrow} b^\dagger_{-k\downarrow} + \mathrm{H.c.}$$

Treat the interband coupling in perturbation theory (to infinite order)

# Interband Pairing

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Odd-interband: $F_{12}^{\text{odd}}(k, i\omega) = \dfrac{F_{12} - F_{21}}{2} = i\omega\Gamma(\Delta_1 - \Delta_2)/D$

Even-interband: $F_{12}^{\text{even}}(k, i\omega) = \dfrac{F_{12} + F_{21}}{2} = \Gamma(\Delta_1\varepsilon_2 - \Delta_2\varepsilon_1)/D$

$$\begin{bmatrix} D = (\omega^2 + E_1^2)(\omega^2 + E_2^2) - \Gamma^2[2(\varepsilon_1\varepsilon_2 - \omega^2) - \Delta_2^*\Delta_1 - \Delta_1^*\Delta_2] + \Gamma^4 \\ E_j^2 = \varepsilon_j^2 + |\Delta_j|^2 \end{bmatrix}$$

Odd-ω pairing: $\Gamma \neq 0, \Delta_1 \neq \Delta_2$

Hybridization and band symmetry breaking

Komendova, Balatsky, and ABS, PRB 92, 04517 (2015)

# ‘Multiband’ Odd-$\omega$ Systems

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- $Sr_2RuO_4$ and $UPt_3$:
  - Multiple bands at Fermi level + Kerr effect → odd-$\omega$, odd-orbital pairing
    Komendova and ABS, PRL 119, 087001 (2017), Triola and ABS, PRB 97, 064505 (2018)

- Doped topological insulator $Bi_2Se_3$
  - Intrinsic bulk SC with odd-orbital nematic order → odd-$\omega$, intraorbital pairing
    Schmidt, Parhizgar, and ABS, PRB (RC) 101, 180512 (2020)

- Bands/Orbitals →
  - Bilayers or TI thin films
    Parhizgar and ABS, SR 7, 9817 (2017)
  - Double nanowires
    Ebisu et al, PTEP 083I01 (2016), Triola and ABS, PRB 100, 024512 (2019)
  - Double quantum dots
    Burset et al, PRB 93, 201402 (2016)
  - Two *k*-space valleys in monolayer TMDs
    Triola et al, PRL 116, 257001 (2016)
  - Pair density wave (PDW) in cuprates
    Chakraborty and ABS, NJP 23, 033001 (2021)
  - Floquet bands in light-driven conventional SCs
    Cayao, Triola, and ABS, PRB 103, 104505 (2021)

# Quasiparticle Interference (QPI)

Direct probe of odd-ω pairing

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# Impurity Scattering

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Impurity scattering changes the local density of states:

$$\rho(\boldsymbol{r},\omega) = \rho_0(\omega) + \delta\rho(\boldsymbol{r},\omega)$$

Fourier Transform:

$$\delta\rho(\boldsymbol{q},\omega) = -\frac{1}{\pi}\operatorname{Im}\left[\sum_{\boldsymbol{k}} \boldsymbol{G}(\boldsymbol{k},\omega)\boldsymbol{T}\boldsymbol{G}(\boldsymbol{k}+\boldsymbol{q},\omega)\right]_{11}$$

**Quasiparticle interference (QPI)**
Fourier transformed STM/STS

Non-magnetic weak impurity: $\boldsymbol{T} = \boldsymbol{V}_{imp}\boldsymbol{\tau}_3$

Nambu Basis: $\boldsymbol{G} = \begin{pmatrix} \boldsymbol{G}_{11} & \boldsymbol{G}_{12} \\ \boldsymbol{G}_{21} & \boldsymbol{G}_{22} \end{pmatrix}$

Anomalous GF: (pair Correlator) $\boldsymbol{G}_{12} = \boldsymbol{F} = \boldsymbol{F}^{e} + \boldsymbol{F}^{o}$

$\boldsymbol{F}^{e}$ ↓ Even; $\boldsymbol{F}^{o}$ ↓ Odd

# Impurities in a SC

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$$\delta\rho(q,\omega) = -\frac{1}{\pi}\,\mathrm{Im}\Big[\sum_k G_{11}(k,\omega)G_{11}(k+q,\omega)$$
$$-F_k^e(\omega)F_{k+q}^e(\omega) - F_k^o(\omega)F_{k+q}^o(\omega)$$
$$-F_k^e(\omega)F_{k+q}^o(\omega) - F_k^o(\omega)F_{k+q}^e(\omega)\Big]$$

Only gap,
no pair correlations

Even in frequency/energy
(set by bias voltage)

Odd in frequency (bias voltage)
Exists only iff odd-ω pairing is present

**→ QPI probes odd-ω pair correlations**

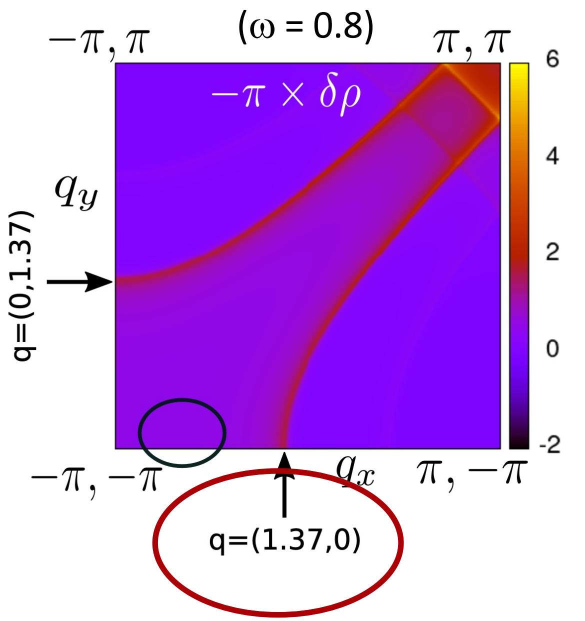
Chakraborty and ABS, PRL 129, 247001 (2022)

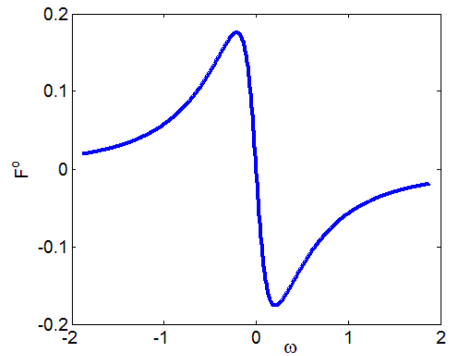
# Simple SC with Odd-ω Pairing

Conventional *s*-wave superconductor + magnetic field

→ **Odd-ω pairing (*s*-wave, spin-triplet)**

QPI at fixed energy in BZ

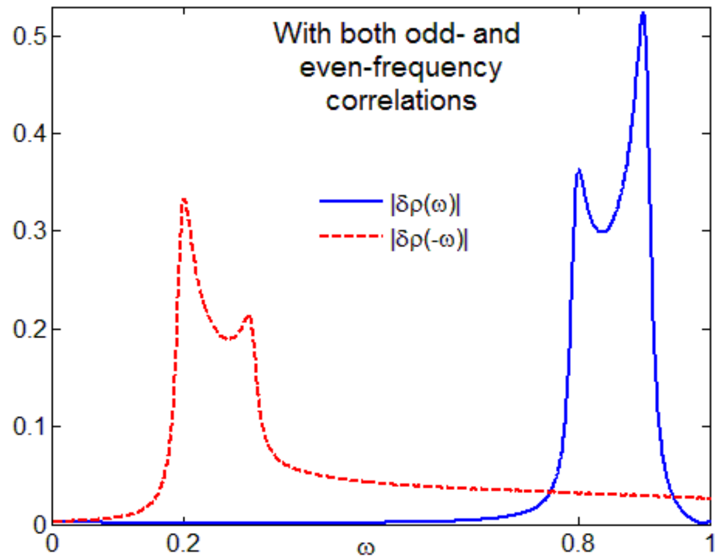


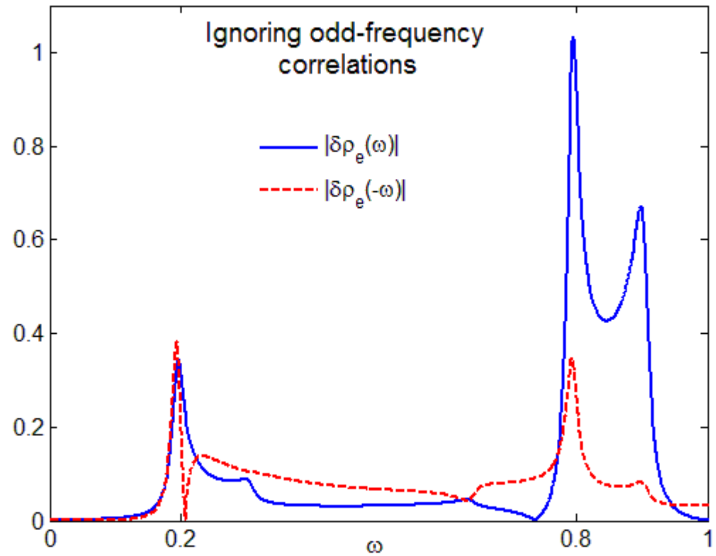



# QPI Bias Asymmetry

At fixed $q$ = (1.37,0)

Forcing $F^{o} = 0$




**Strong bias peak *position* asymmetry due to odd-ω pairing**

(Peak shapes set by normal-state properties)

Chakraborty and ABS, PRL 129, 247001 (2022)

# QPI Bias Asymmetry

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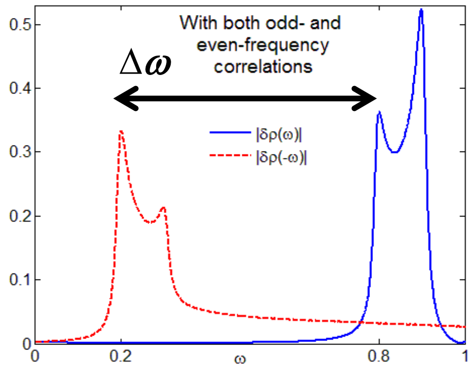
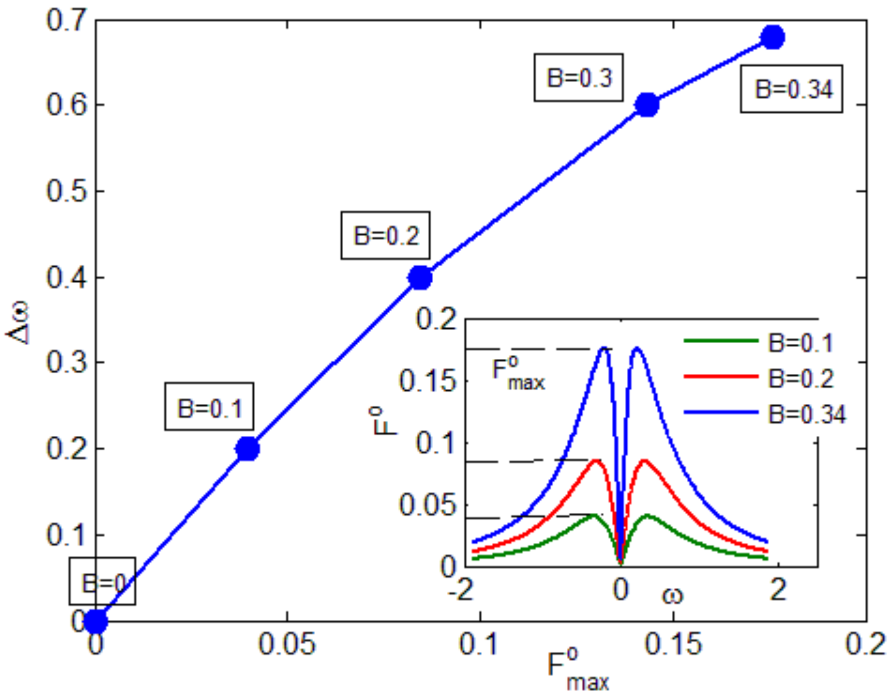
Peak distance




**Odd-ω pairing ∝ peak distance**

Chakraborty and ABS, PRL 129, 247001 (2022)

# Summary – Odd-ω Superconductivity

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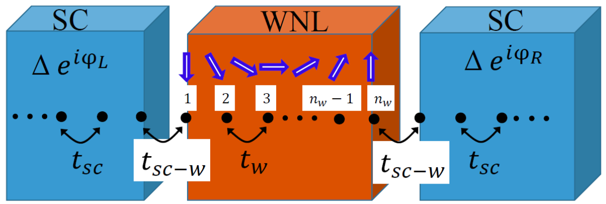
- Dynamic & hidden SC pair correlations

$$\Delta \sim F = \langle \psi_\alpha(t)\psi_\beta(0) \rangle$$

- Weyl nodal loop semimetals make optimal odd-ω Josephson junctions




- Multiband systems: **SPOT = -1**
  - Multi-band/orbital/layer/dot/wire/valley/momenta/Floquet bands/…

- QPI probes odd-ω pair correlations